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DEMUR: Double Electron Muon Resonance

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This paper will present the details of a general theory of DEMUR, with examples of current experimental interest: quartz, where the hyperfine interaction is nearly isotropic; and silicon, wherein an anisotropic species has been found. The theory will be compared with experimental data.

Electron paramagnetic resonance (EPR) transitions driven by an intense uniform radio-frequency (rf) magnetic field result in characteristic structure in the frequency spectrum of time-differential μSR data for muonium-like centers in nonmetals. This phenomenon is labeled Double Electron Muon Resonance (DEMUR). The theory of DEMUR involves solving the time-dependent Schrödinger Equation neglecting relaxation, an approximation which is valid for muonium in quartz at room temperature where the first observations of DEMUR were made[1]. This assumption would also appear to be valid for anomalous muonium, Mu^*, in silicon, germanium, and diamond at sufficiently low temperatures. The hyperfine interaction for muonium in quartz is nearly isotropic and large whereas that for Mu^* is very anisotropic and small. The theory of DEMUR for both cases will be discussed and the isotropic case will be compared to observations in quartz.

The analysis proceeds by writing the spin Hamiltonian in terms of the total magnetic field which contains both the constant field and the time-dependent field. Quite generally for an electron spin of $\frac{1}{2}$ and excluding any nuclear spins we have

$$\mathcal{H} = \mu_B \vec{H}_t \cdot \overleftrightarrow{g} \cdot \vec{S} + \vec{S} \cdot \overleftrightarrow{A} \cdot \vec{I} - g_\mu \mu_\mu \vec{H}_t \cdot \vec{I} \qquad (1)$$

where the total magnetic field includes a uniform linearly-polarized rf field,

$$\vec{H}_t = \vec{H} + \vec{H}_1 \cos(\omega t + \phi). \qquad (2)$$

Since in our calculation the muon stops at $t = 0$, ϕ is the phase angle of the rf field at this time.

The stationary states of the time-independent spin Hamiltonian have wave functions ψ_n and energies $\hbar\omega_n$ with $n = 1,\ldots,4$. The time dependent wave function can then be written as

$$\psi(t) = \sum_{n=1}^{4} g_n(t)\, \psi_n\, e^{-i\omega_n t} \tag{3}$$

in terms of the time-dependent coefficients $g_n(t)$. The initial values of the $g_n(t)$ are determined by specifying the initial state of the system. In this initial state the muon spin state is specified (it is 100% polarized), but the electron state is random.

Substituting the expression in Eq. 3 for $\psi(t)$ into the time-dependent Schrödinger Equation, including the time dependent field as in Eq. 2, allows four coupled differential equations for the $g_n(t)$ to be written:

$$i\frac{dg_m}{dt} = \frac{1}{\hbar}\sum_{n=1}^{4} g_n e^{i\omega_{mn}t}\langle\psi_m|\mu_B\vec{H}_1\cdot\overleftrightarrow{g}\cdot\vec{S} - g_\mu\mu_\mu\vec{H}_1\cdot\vec{I}|\psi_n\rangle \cos(\omega t + \phi), \tag{4}$$

where $\omega_{mn} = \omega_m - \omega_n$. The solution of Eq. 4 gives the time-dependent wave function from which we can calculate the components of the muon spin polarization. In terms of the $g_n(t)$ these are given by

$$\vec{P} = 2\sum_{m,n=1}^{4} g_m^*(t)g_n(t)\, e^{i\omega_{mn}t}\, \langle\psi_m|\vec{I}|\psi_n\rangle. \tag{5}$$

The coupled differential Eqs. 4 suggest a very complicated time dependence for the $g_n(t)$. However, the only terms on the right side of Eq. 4 which lead to a contribution to the $g_n(t)$ with appreciable amplitude are those at low frequency. In the simplest approximation the rf frequency is on or near resonance with a single transition frequency $\omega_{k\ell}$, which is well-separated from any other transition frequency of the system. Then the differential equations simplify considerably. Taking $\omega \approx \omega_{k\ell} > 0$ and labeling the other two stationary states by m and n we have

$$\frac{dg_m}{dt} = \frac{dg_n}{dt} = 0$$

$$i\frac{dg_k}{dt} = \frac{1}{2\hbar}\, g_\ell\, e^{-i(\omega-\omega_{k\ell})t}\, e^{-i\phi}\langle\psi_k|\mu_B\,\vec{H}_1\cdot\overleftrightarrow{g}\cdot\vec{S} - g_\mu\mu_\mu\,\vec{H}_1\cdot\vec{I}|\psi_\ell\rangle,$$

$$i\frac{dg_\ell}{dt} = \frac{1}{2\hbar}\, g_k\, e^{i(\omega-\omega_{k\ell})t}\, e^{i\phi}\langle\psi_\ell|\mu_B\,\vec{H}_1\cdot\overleftrightarrow{g}\cdot\vec{S} - g_\mu\mu_\mu\,\vec{H}_1\cdot\vec{I}|\psi_k\rangle. \tag{6}$$

Eqs. 6 require that $g_m(t)$ and $g_n(t)$ be constant. In solving Eqs. 6 it is convenient to write

$$h_\ell(t) = g_\ell(t)\, e^{-i\phi} \tag{7}$$

and to define

$$\omega_{k\ell} = \frac{1}{\hbar}\langle\psi_k|\mu_B\vec{H}_1\cdot\overleftrightarrow{g}\cdot\vec{S} - g_\mu\beta_\mu\,\vec{H}_1\cdot\vec{I}|\psi_\ell\rangle. \tag{8}$$

Then Eqs. 6 become

$$i\frac{dg_k}{dt} = \frac{1}{2}\,\omega_{k\ell}h_\ell\, e^{-i(\omega-\omega_{k\ell})t},$$

$$i\frac{dh_\ell}{dt} = \frac{1}{2}\,\omega^*_{k\ell}g_k\, e^{i(\omega-\omega_{k\ell})t}. \tag{9}$$

The solutions of Eqs. 9 still involve a moderate amount of algebra; however, we can see several features of the solutions. Since the equations are coupled and the initial value of $g_k(t)$ is independent of ϕ whereas the initial value of $h_\ell(t)$ is $g_\ell(0)e^{i\phi}$, we have two terms in the solutions, one independent of phase and one proportional to $e^{i\phi}$. In addition, both $g_k(t)$ and $h_\ell(t)$ will be oscillatory with two frequency components. The solutions, written in terms of g_k and g_ℓ, are

$$g_k(t) = \left[\frac{1}{2}\left(1 + \frac{z}{\sqrt{z^2+1}}\right)g_k(0) - \frac{1}{2\sqrt{z^2+1}}g_\ell(0)\frac{\omega_{k\ell}}{|\omega_{k\ell}|}e^{-i\phi}\right]e^{-i\omega_- t}$$
$$+ \left[\frac{1}{2}\left(1 - \frac{z}{\sqrt{z^2+1}}\right)g_k(0) + \frac{1}{2\sqrt{z^2+1}}g_\ell(0)\frac{\omega_{k\ell}}{|\omega_{k\ell}|}e^{-i\phi}\right]e^{-i\omega_+ t},$$

$$g_\ell(t) = \left[\frac{1}{2}\left(1 - \frac{z}{\sqrt{z^2+1}}\right)g_\ell(0) - \frac{1}{2\sqrt{z^2+1}}g_k(0)\frac{|\omega_{k\ell}|}{\omega_{k\ell}}e^{i\phi}\right]e^{i\omega_+ t}$$
$$+ \left[\frac{1}{2}\left(1 + \frac{z}{\sqrt{z^2+1}}\right)g_\ell(0) + \frac{1}{2}\frac{1}{\sqrt{z^2+1}}g_k(0)\frac{|\omega_{k\ell}|}{\omega_{k\ell}}e^{i\phi}\right]e^{i\omega_- t}, \tag{10}$$

where $$\omega_\pm = \frac{1}{2}\left(z \pm \sqrt{z^2+1}\right)|\omega_{k\ell}| \tag{11}$$

and $$z = \frac{\omega - \omega_{k\ell}}{|\omega_{k\ell}|}. \tag{12}$$

This leads to three different kinds of results when Eq. 10 and the constant values of $g_m(t)$ and $g_n(t)$ are introduced into Eq. 5. First, if the frequency $\omega_{k\ell}$ is observed as a muon precessional frequency, then it will be split into three frequency components, the middle frequency appearing at the rf driving frequency ω flanked by two lines displaced in frequency by $\sqrt{z^2+1}\,|\omega_{k\ell}|$. If the phase angle ϕ is random, as it was in the first observations made on quartz[1], then the amplitude of the three lines (lowest frequency to highest) are $\frac{1}{4}\left(1 - \frac{z}{\sqrt{z^2+1}}\right)^2$, $\frac{1}{2}\frac{1}{z^2+1}$, and $\frac{1}{4}\left(1 + \frac{z}{\sqrt{z^2+1}}\right)^2$ times the amplitude with no rf. Second, if the observed muon precessional frequency involves either the k or ℓ state, but not both, then the normal µSR frequency is split into two lines with a splitting of $\sqrt{z^2+1}\,|\omega_{k\ell}|$. Again for random phase angle ϕ, the amplitude of the lower frequency line is $\frac{1}{2}\left(1 - \frac{z}{\sqrt{z^2+1}}\right)$ times the amplitude with no rf and the amplitude of the higher frequency line is $\frac{1}{2}(1 + \frac{z}{\sqrt{z^2 + 1}})$ times the no rf amplitude if $\omega_k > \omega_m$ or $\omega_m > \omega_\ell$ (recall that $\omega_k > \omega_\ell$) and the sign of z is reversed otherwise. Finally, if ω_{mn} is observed as a muon precessional frequency, then no splitting occurs.

These results are quite general. They do not depend upon the complexity of the spin Hamiltonian nor upon the details of the energy levels or wave functions. They depend only upon having $\omega_{k\ell}$ removed from any strong magnetic-dipole transition by several times the $|\mu_{ij}|$ of these transitions. The details of a specific system enter in only when determining $|\mu_{k\ell}|$ and the no rf frequencies, amplitudes, and phases.

EXPERIMENTAL RESULTS

For quartz only two μSR lines exist at reasonable frequencies; by applying an rf field near one of these two transition frequencies the most important splittings characteristic of DEMUR are observed. A representative portion of such experimental data on quartz is contained in Table 1. This data was taken with a field of 124.7 Gauss, where the μSR lines appear at 167.2 MHz and 180.5 MHz. The spectra presented were each analyzed by a least squares fit to any frequency components observed in a Fourier transform. The applied field amplitudes quoted are determined from the splitting in the DEMUR results.

The first set of data shows a DEMUR spectrum with the rf frequency near the lower μSR transition (167.2 MHz). The three components of this spectrum with frequencies closest to that of the lower μSR line are characteristic of the transition being irradiated by the rf field also appearing in the μSR. The other two components, appearing near the upper μSR frequency, show the characteristics of the observation of a transition sharing only a single eigenstate with the irradiated transition. The second set of data was obtained with the rf frequency near the upper μSR transition (180.5 MHz), reversing the roles of the two μSR transitions from that found in the first data set.

Table 1: Results of DEMUR on Quartz

Applied Field	Experimental Results Freq. (MHz)	ampl.	Theory* Freq.	ampl.
167.301	166.25(3)	.0133(9)	166.32	.0124
MHz	167.24(2)	.0187(9)	167.30	.018[illegible]
1.0 G.	168.17(5)	.0064(9)	168.28	.007[illegible]
	179.69(3)	.0098(9)	179.92	.009[illegible]
	180.84(3)	.0100(9)	180.90	.010[illegible]
180.510	166.56(3)	.0115(10)	166.53	.0115
MHz	167.68(3)	.0139(10)	167.68	.0139
1.0 G.	179.91(4)	.0081(10)	179.47	.008[illegible]
	180.42(3)	.0141(10)	180.51	.014[illegible]
	181.51(5)	.0061(10)	181.55	.0061

* The theoretical amplitudes are normalized to give the observed total amplitude within each group of lines.

REFERENCE

[1]J A Brown, et. al., Phys. Rev. Lett. 43, 1751 (1979).